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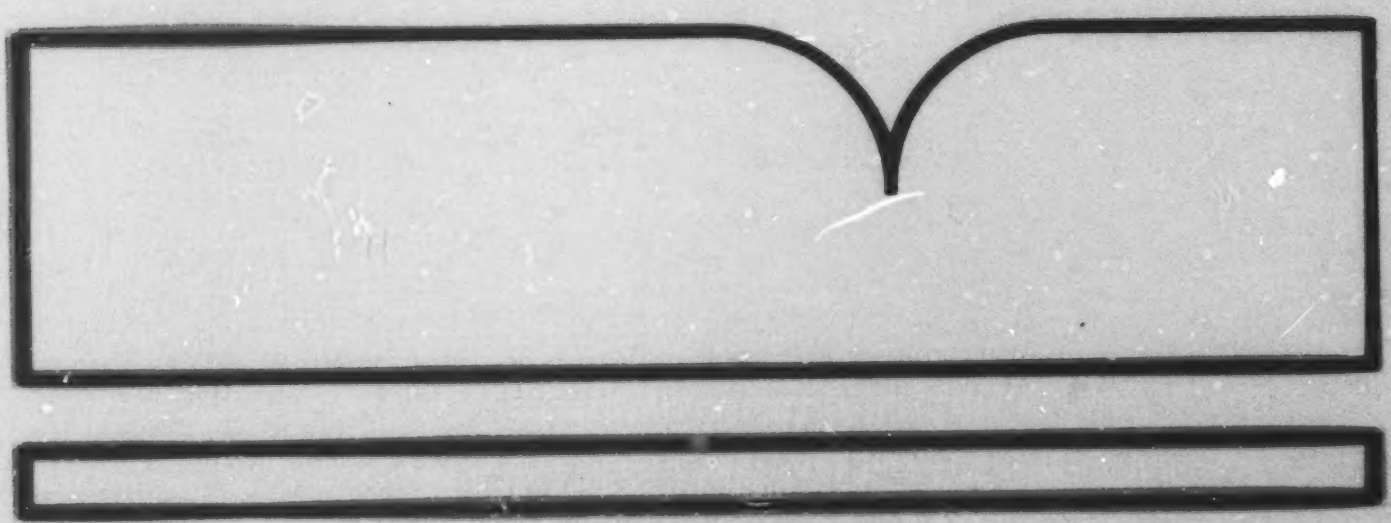

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U.S./GERMAN LIMB TECHNOLOGY TRANSFER

by

J.L. Reese and R. Payne
Energy and Environmental Research Corporation
18 Mason
Irvine, California 92718

and

Y. Chughtai
L&C Steinmuller GmbH
Federal Republic of Germany

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Project Officer

David G. Lachapelle
U.S. Environmental Protection Agency
Air and Energy Engineering Research Laboratory
Research Triangle Park, North Carolina 27711

AIR AND ENERGY ENGINEERING RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
RESEARCH TRIANGLE PARK, NC 27711

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a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group	
Pollution	Calcium Hydroxides	Pollution Control	13B	
Utilities	Fly Ash	Stationary Sources		21B
Boilers	Coal	Utility Boilers	13A	21D
Nitrogen Oxides	Particles	Limestone Injection Multistage Burners (LIMB)	07B	14G
Sulfur Oxides	Electrostatic Precipitators			
Limestone		Staged Burners	08G	31I

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ABSTRACT

This report presents key findings of a program in which the U.S. EPA participated in a program sponsored by the Umweltbundesamt (UBA), the German equivalent of the EPA. The scope of the UBA program included retrofitting the 700 MWe Weiher III utility boiler of the Saarbergwerke AG with staged-mixing burners for NOx control, and sorbent injection for SOx control. This program was considerably reduced in scope because of restrictions placed upon the utility by local environmental officials concerning the classification of the fly ash generated in the process. During the limited testing period, SO_2 emissions were reduced 8 to 64 percent depending on Ca/S molar ratio and other operating conditions. The program originally planned at Weiher was ultimately conducted at the 48 MWe Tiefstack Unit 6 of Hamburgishe Elektrizitats-Werke AG. Moderate levels of SO_2 control were achieved (22 percent with limestone, and 43 percent with calcium hydroxide). An analysis of the test results suggests that the use of more reactive sorbents could increase the SO_2 removals to 30 percent and 60 percent at a Ca/S ratio of 2, for limestone and calcium hydroxide, respectively.

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BACKGROUND

Control of SOx emissions through dry sorbent injection into boiler furnaces was extensively explored in the U.S. during the late 1960's and early 1970's with limited success. Typically, SOx removal efficiencies of 18 to 40 percent were achieved for a variety of boilers over a wide range of calcium-to-sulfur (Ca/S) molar ratios. At economic Ca/S ratios of 1.3 to 2.0, sulfur capture was typically 20 to 33 percent respectively. These lower (than expected) removals were attributed to: deadburning of sorbent, and decomposition of the resultant calcium-sulfur complex at high temperatures, and; inadequate mixing (contacting) of the sorbent with the combustion gases. These results were not competitive with the high (80 to 90 percent) sulfur oxide removal efficiencies of wet scrubbers, and consequently, further work with dry sorbents was abandoned around 1973.

More recently, attention refocused on dry sorbent SOx control technology. Prompting this was: (1) a reexamination of reaction chemistry that indicated differences from what was previously accepted; (2) new combustion conditions and mixing patterns that appeared to favor sorbent injection in a manner different than previously employed; (3) a need for low-cost NOx and SOx control technology to support acid rain control strategies, and; (4) pilot-scale tests that showed improved SOx capture with more reactive sorbents. The EPA acronym for this low-NOx/SOx approach is LIMB (Limestone Injection Multistage Burner). EPA-sponsored development programs were underway, and at the time of this project, significant work was in progress in the Federal Republic of Germany.

One of the major objectives of the LIMB program is to assess the impact of LIMB technology on utility boiler design. This is a critical element, and it is recognized that the utility boiler (burners, furnace, radiant and convective sections) and ancillaries (particulate cleanup, ash/spent product removal, etc.) need to be carefully evaluated as to the implications of future LIMB applications for both retrofit and new unit designs.

In this project, the U.S. EPA participated in a program sponsored by the Umweltbundesamt (UBA), the German equivalent of the EPA. The scope of the UBA program included retrofitting the 700 MWe Weiher III utility boiler of the Saarbergwerke AG with staged-mixing burners for NOx control, and sorbent injection for SOx control. The retrofit was performed by L&C Steinmuller GmbH, a major German boiler manufacturer. The program was to be the first full-scale evaluation of LIMB technology and presented a unique opportunity for EPA to participate in the program at relatively modest cost. All data developed in the program would be shared between the Federal Republic of Germany and the United States. EPA's participation was essentially concerned with

emissions monitoring and characterization of particulate, slagging and fouling, and determination of gas and particle temperatures which are critical parameters to the successful application of LIMB technology. Additionally, this program would augment on-going EPA-sponsored development programs and design criteria studies, and provide an essential link between EPA pilot- and prototype- scale data and full-scale data in the U.S.-sponsored LIMB development effort.

SUMMARY OF SORBENT INJECTION TESTS AT WEIHER III

Weiher III is a 700 MWe coal-fired unit operated by the German utility Hamburgishe Elektrizitats-Werke AG (HEW). The furnace is divided into two combustion chambers, and is equipped with a total of 24 staged-mixing burners. Twelve burners each are installed in the front and rear walls in three elevations by four burners horizontally. Groups of four burners are each served by one mill, and are suppled with sorbent through an injection system. Sorbent is transported by compressed air.

A pre-condition to testing at Weiher was that the boiler was not to be put at risk in any way. Initially, only two burners on the middle plane of the left combustion chamber were modified to investigate potential operating problems. The objective of this first phase was primarily to investigate fouling in the immediate area of the burner, in the furnace, and in the convective section, as well as to determine alterations to the ash characteristics.

The results of tests with two burners in 1982 were good enough to provide the recommendation to equip all 24 burners of the boiler with sorbent injection. These tests were started after considerable delay, mainly due to the classification of the fly ash as "hazardous waste" by the Landesamt fur Umweltschutz und Wasserwirtschaft --LFU (Office for Environmental Protection). The following steps were taken in order to comply with the LFU conditions:

Provide an overheat prevention wetting system for the ash conveyor belt to slake free CaO in the fly ash.

Provide for intermediate storage of fly ash in a standby settling basin.

Provide for neutralization of the wet-ash removal water.

Transport ash to a special dump for hazardous wastes in Schonenberg (German Democratic Republic).

The baseline tests without sorbent injection were performed in August 1984, wherein data were collected regarding naturally

occuring sulfur capture (due to alkaline material in the coal ash), ash characteristics, and process data for the boiler, air heater, and electrostatic precipitator.

The originally planned program of sorbent injection tests had to be considerably reduced due the restrictions imposed relative to ash disposal. Abbreviated tests were planned with $CaCO_3$ and $Ca(OH)_2$ at different boiler loads, Ca/S molar ratios, and injection planes. These tests were scheduled to take three days for each of the additives involved.

After receipt of official approval for the execution of the sorbent injection tests in November 1984, the tests were immediately started. The first sorbent used was pulverized limestone. It was observed during the course of these tests that the 4:1 molar ratio required at full boiler load could not be attained. The problem appeared to be related to the ejector design. A maximum flow corresponding to a Ca/S of 2.5 was achievable. However, when the six injection lines were run in parallel for continuous feed to all burners, the maximum possible limestone flow was found to be equivalent to a Ca/S molar ratio of only about 1.4. Full load tests with sorbent to all burners were conducted at molar ratios between 0.5 and 1.4. Due to the reduced rate, there was a wide variation in SO_2 capture results. Those obtained only indicated a general trend. Calcium utilization decreased with increasing molar ratio and was on the order of 10 to 20 percent. Typical SO_2 captures at full load were in the range of only 8 to 12 percent, at Ca/S ratios of 0.7 and 1.4 respectively. Limited tests were run at 60 percent load, and Ca/S ratios of 2.5 and 4.0 were achievable. An improvement in both calcium utilization and SO_2 capture was observed. At Ca/S of 2.5, the calcium utilization was 11 percent with an SO_2 capture of about 28 percent. At Ca/S of 4, the calcium utilization was 16 percent, with an SO_2 capture of 64 percent. No effects attributable to variation on the injection point (upper or lower burner plane) on the calcium utilization could be ascertained.

Between sootblower operation, there was a tendency for furnace temperatures to rise during sorbent injection. The maximum temperature increase of 30 Centigrade degrees at full load was measured with the highest sorbent flow at Ca/S of 1.4. With sorbent injection, unburned combustibles in the fly ash fell from the normal level of 1.5 to 3 percent to a value of less than one percent. This corresponds to the increased furnace temperatures.

Continuously increasing fouling was found on all walls in the areas of the middle and upper burner planes between sootblowing operating cycles during sorbent injection. The first convective heating surfaces also showed more pronounced fouling. The deposits, due to their high lime concentration, were easily removed by sootblowing. This was confirmed by inspection of the

boiler after completion of the tests and also by records of the superheater and reheater spray flows before and after sootblowing. No increase in fouling was evident for the air heater. The pressure drop across the air heater was constant over the entire test period.

After completion of the 3-day tests with limestone, the electrostatic precipitator (ESP) was inspected. Heavier than normal dust deposits were found on the discharge electrodes and plates in the middle and the final cleaning zones of the ESP. These deposits were sticky, and had a high fines content; i.e., 66 percent of the material was smaller than 8 microns. The material also had high sulfur content. The deposits could not be removed through normal blowing and rapping. The higher fouling tendency in the middle and final precipitator stages was attributed to the higher fines content in these stages resulting from sorbent injection. This higher dust loading, and the increase in fly ash resistivity, produced a higher load on the middle and final precipitator stages.

The propagation of deposits on the precipitator surfaces, which could not be removed by normal means, was a key factor in the boiler owners decision to discontinue further sorbent injection tests. The owners fear that the fouling phenomena would intensify if the tests were continued, and that the deposits could not be removed without damage to the precipitator could not be assuaged to their full satisfaction.

These circumstances, together with the very expensive arrangements that had to be made for fly ash disposal, resulted in premature cessation of the tests. The scheduled tests with calcium hydroxide were expected to lead to a better utilization of calcium but could not be carried out.

The Weiher III operating staff have meanwhile made several efforts to clean the precipitator. By switching off paths in the preliminary and middle precipitator stages, they have reduced deposits in the final stage areas. A recent inspection of the precipitator surfaces has shown that there are no deposits left at all.

The planned measurement program for EPA that was originally scheduled for the Weiher power station was later performed at the HEW Tiefstack power station during November and December 1985.

SUMMARY OF SORBENT INJECTION TESTS AT TIEFSTACK

Furnace sorbent injection to control SO_2 emissions at the Tiefstack power station is being considered by the utility owner HEW, following encouraging results on other units. A temporary

injection system was installed by HEW on Boiler 6 to evaluate the process. HEW plans ultimately to install permanent sorbent injection systems on all six boilers at the Tiefstack station. The application of sorbent injection to Boiler 6 was evaluated by the Energy and Environmental Research Corporation (EER) as a subcontractor to L&C Steinmuller, GmbH, for the U.S. EPA. The evaluation involved field measurements at Tiefstack during injection tests and the use of models of the sulfation process to aid in the interpretation of the test results. The objectives of the program were to use the field test results to directly evaluate the effects of sorbent injection on SO_2 emissions and boiler performance, to evaluate the valididty of the process models, and to use the models to identify optimum injection parameters and estimated maximum SO_2 reductions.

Figure 1 shows the procedure used to evaluate sorbent injection with the process models. The in-furnace sulfation reaction is dependent on four key parameters:

1. Gas temperature at the injection location
2. Furnace gas temperature quench rate
3. Sorbent reactivity
4. Sorbent disperaion in the furnace

A three dimensional heat transfer model of the boiler is used to determine the furnace gas temperature distribution. This then allows identification of the injection location having the optimum furnace gas temperature of about 1530K (2300 ^{o}F), and the temperature quench rate. The quench rate determines the residence time of the gas in the sulfation reaction temperature range of approximately 1530 to 1250K (2300 to 1800 ^{o}F). Sorbent reactivity is determined by injection of the sorbent into a bench scale furnace. EER has evaluated a broad range of sorbents to determine relative reactivities. Disperaion of the sorbent in the furnace is evaluated in a small-scale isothermal flow model. A range of visualization and tracer techniques are used to determine furnace gas flow patterns. Sorbent injection configurations are evaluated using injection of smoke or a tracer gas. A mathematical model of the sulfation reaction may then be used to evaluate sulfur capture based on the injection temperature, quench rate, sorbent reactivity, and disperaion. The model can be used to predict capture with alternate sorbents, injection locations, or dispersion patterns for the boiler of interest, to determine the system design providing the highest capture. The model may also be used to extrapolate the test results to boilers of different design.

BOILER DESCRIPTION

The boiler evaluated in the program, Tiefstack Unit 6, was constructed in 1952 by Durr Werke AG. The boiler is tangentially fired with low-sulfur bituminous coal. The steam flow

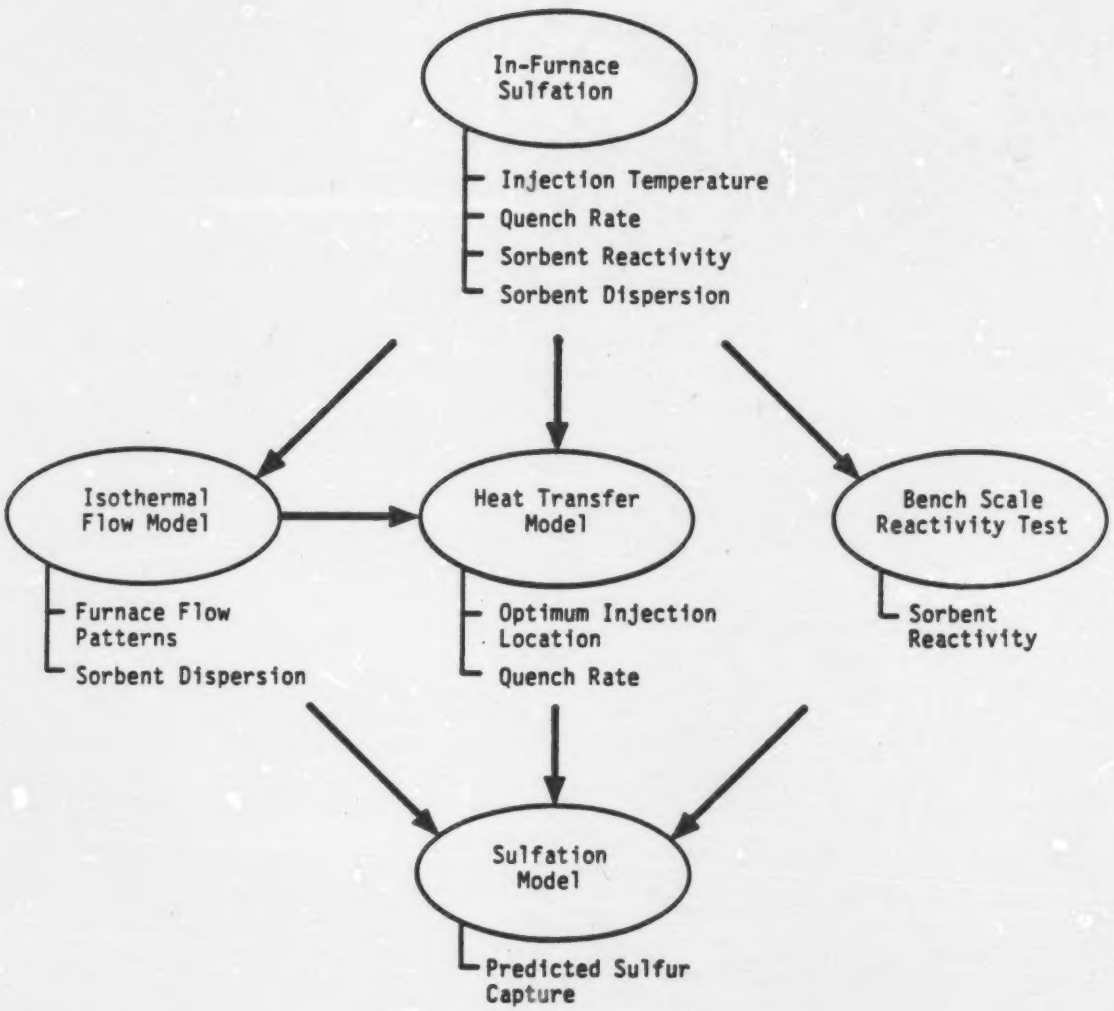


Figure 1. Sorbent injection evaluation procedure.

configuration is once-through, with the steam used for electrical generation and district heating. The maximum rated steam flow is 160,000 kg/hr (352,000 lb/hr) at 783K (950 °F) and 140 bar (2000 psig). Figure 2 shows a diagram of the boiler with injection locations in existing wall ports.

In this boiler, the radiant superheater consists of wall tubes rather than pendant tubes. This design significantly increases the furnace volume prior to the convective section, resulting in a lower gas quench rate. The convective section consists of superheater, reheater, and economizer passages. The unit has a tubular air heater, with an electrostatic precipitator (ESP) for fly ash collection.

The temporary sorbent injection system was installed and operated by the HEW utility. Sorbent was fed from pneumatic trucks to a small feed hopper located inside the boiler house, and then fed into a pneumatic transport line with a rotary feeder and ejector. Air for transport and injection was supplied by a rotary air blower. Injection velocity was varied by changing the transport air flow rate, or by varying the nozzle diameter. Due to limitations of the injection system design, and the use of existing observation ports, sorbent was injected through only one or two nozzles using available ports on the boiler.

The time/temperature profile of the furnace gas was calculated using a three-dimensional heat transfer model. This profile is shown in Figure 3. Figure 4 shows the furnace gas temperature profile with boiler elevation. Accuracy of the model calculations was confirmed by measurements during the field evaluation, as shown in the figure. Model predictions and measurements confirm that the injection location used in the field test was near optimum, with a mean furnace gas temperature of 1520K (2280 °F). The quench rate in the upper furnace was 140K/sec (250 F°/sec), with a residence time of about 2.5 seconds in the sulfation temperature window. This quench rate is much lower than typical U.S. boilers where 450K/sec (800 F°/sec) is not unusual. The low quench rate at Tiefstack thus allows a relatively long time for sorbent/furnace gas mixing and for the sulfation reaction. Measurements at reduced boiler load showed the gas temperature to be reduced to 1440K (2130 °F) at the injection elevation.

FIELD TEST RESULTS

Field testing was performed from November 29 through December 13, 1985. Figure 5 shows the SO_2 reductions obtained as a function of Ca/S ratio. Injection parameters varied during the test included:

- Number of nozzles (one or two)
- Injection velocity

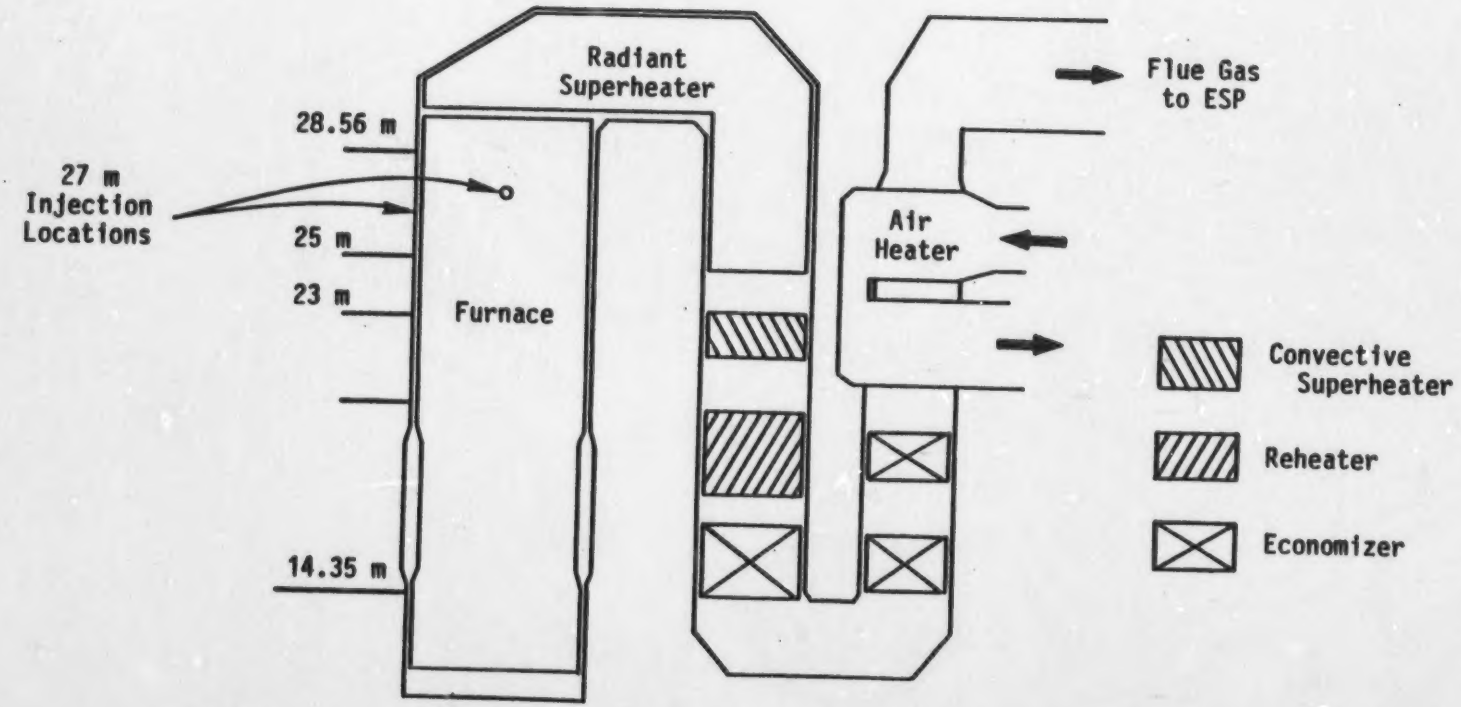


Figure 2. Tiefstack Boiler 6.

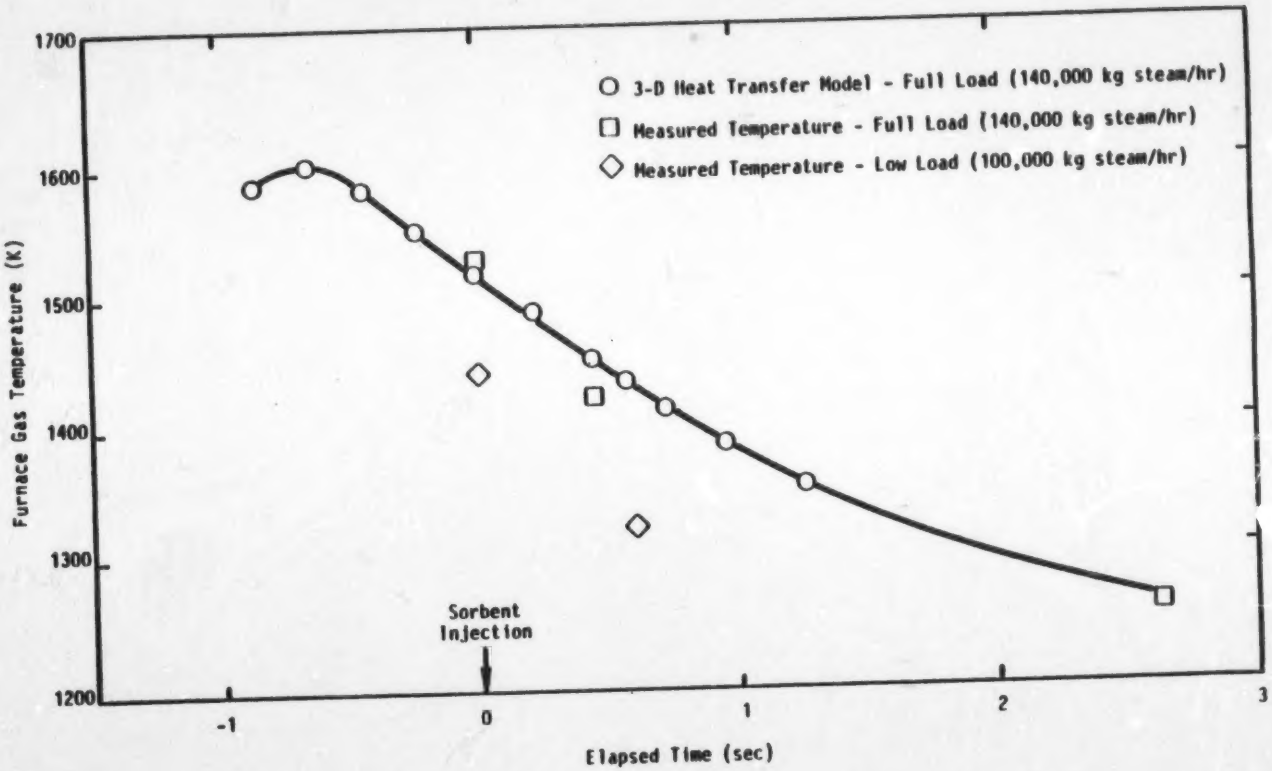


Figure 3. Tiefstack time/temperature profile.

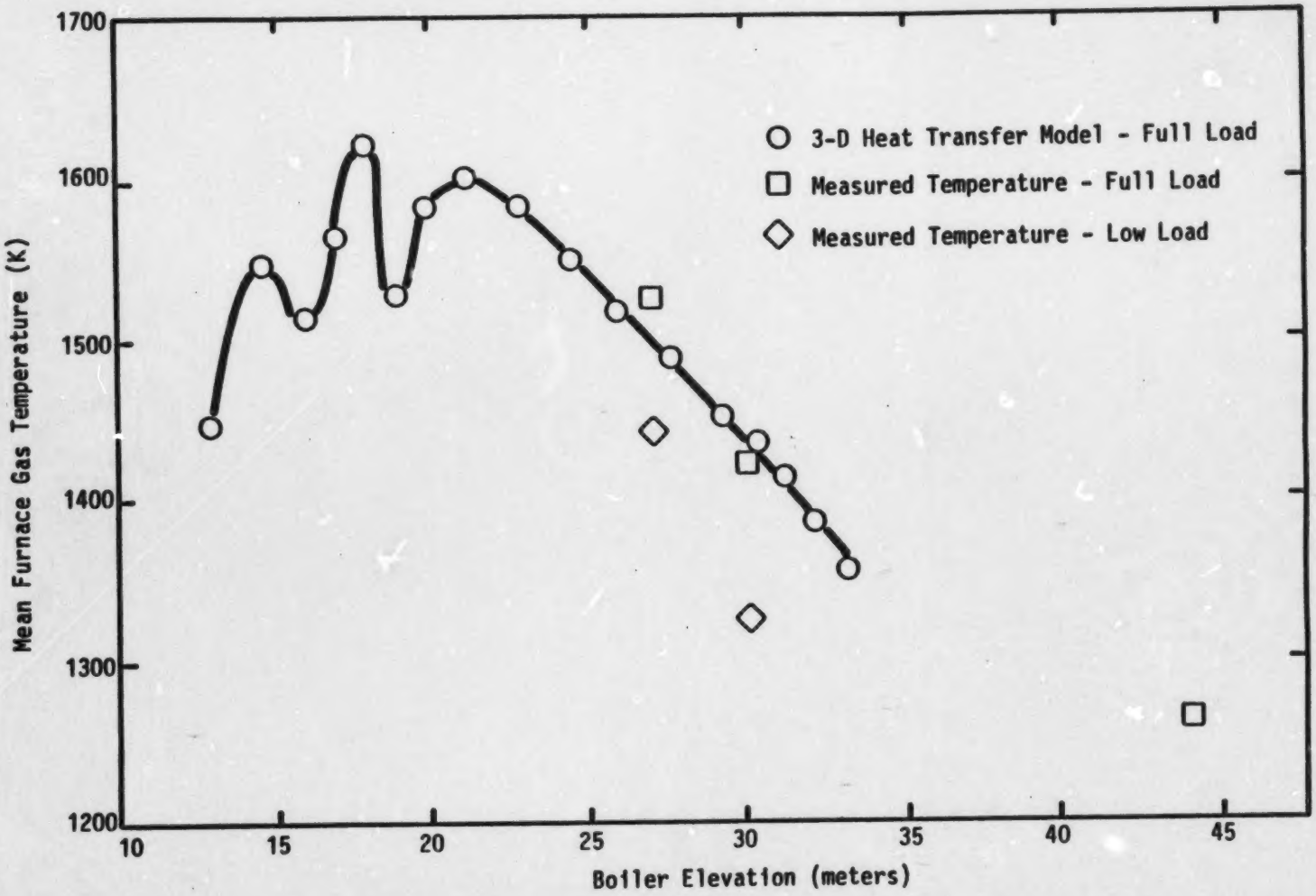
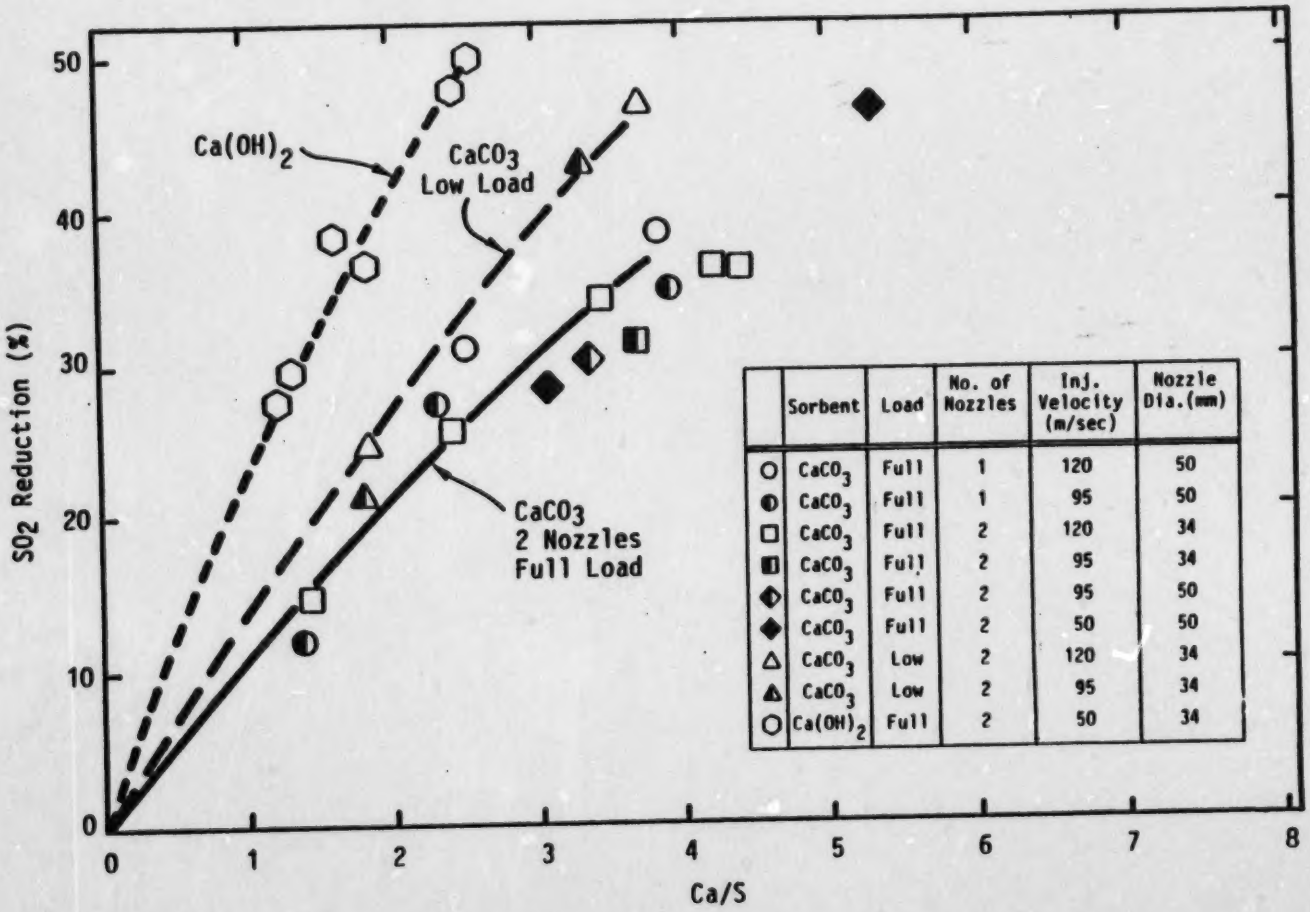


Figure 4. Temperature vs. boiler elevation at Tiefstack.



	Sorbent	Load	No. of Nozzles	Inj. Velocity (m/sec)	Nozzle Dia.(mm)
○	$CaCO_3$	Full	1	120	50
◐	$CaCO_3$	Full	1	95	50
□	$CaCO_3$	Full	2	120	34
◧	$CaCO_3$	Full	2	95	34
◈	$CaCO_3$	Full	2	95	50
◆	$CaCO_3$	Full	2	50	50
△	$CaCO_3$	Low	2	120	34
▲	$CaCO_3$	Low	2	95	34
⬡	$Ca(OH)_2$	Full	2	50	34

Figure 5. Tiefstack - field results.

- Boiler load
- Sorbent type (limestone or hydrated lime)

For injection with one nozzle, the injector was located on the front wall of the boiler, off-center to the right, resulting in injection countercurrent to the furnace gas flow. For two-nozzle injection, a second injector was added at the center of the right-hand wall, resulting in injection normal to the furnace gas flow.

Changes in sorbent disperaion patterns resulting from variations in the number of injectors or injection velocity, resulted in little change in SO_2 removal rates. Sorbent utilization appears to be equal or slightly higher with one injector compared to two. Increasing sorbent penetration by increasing the injection velocity resulted in a slight improvement in results. Reducing boiler load was the only parameter that resulted in a significant change in capture. With a decrease in load to 71 percent of full load during limestone injection, sulfur removal increased from 22 to 27 percent at a Ca/S ratio of 2.0, corresponding to an increase in calcium utilization from 11.0 to 13.5 percent. The improved results with reduced load are believed to be due to increased sorbent reactivity associated with a lower furnace gas temperature. Injection of a higher reactivity sorbent, atmospheric calcitic hydrate (hydrated lime), resulted in a corresponding increase in SO_2 removal, yielding a calcium utilization of 21.5 percent at a Ca/S ratio of 2.0, and full load. All SO_2 removal data were verified by solids sampling and wet-chemical SO_2 determinations conducted during the tests.

Sorbent injection did not result in any noticeable impacts on boiler performance. Over a 24 hour test period, no changes in heat absorption were seen, as evidenced by steam temperatures, attemperation flow rates, or sootblowing requirements. A normal sootblowing frequency of once each shift was maintained throughout the injection tests. Measurements of furnace radiation using an ellipsoidal radiometer did not show any change with sorbent injection. Some slag buildup was noted in the lower portion of furnace in the burner zone. This buildup was typical of normal operation, however, and not believed to be caused by the sorbent. The upper furnace walls and convective passage did not experience any slagging or fouling during the test. The sorbent injection also did not necessitate any changes in procedures to remove ash from the boiler or the ESP. Evaluation of precipitator performance was not part of this program, and no particulate loading measurements were performed at the precipitator outlet. A maximum increase in opacity of less than 5 percent above the baseline of 10 percent was noted at the precipitator outlet during sorbent injection. Effects of the sorbent on precipitator performance were offset, however, by the high carbon content of the ash at Tiefstack. A minimum carbon content of 12 percent is specified by the purchaser of the fly

ash. This carbon content controls the resistivity of the fly ash and overrides the impact of the sorbent.

ANALYSIS OF TEST RESULTS

Reductions of SO_2 obtained during the field test were somewhat lower than anticipated, based on the low quench rate of the boiler. Field measurements confirmed that the injection temperature was near optimum. Subsequent evaluations therefore focused on the dispersion of the sorbent and sorbent reactivity.

The effectiveness of sorbent dispersion was evaluated in a small scale isothermal flow model. A qualitative assessment of mixing was made using smoke injection. These results showed the flow of the injection jets to be dominated by the furnace gas flow where the sorbent jets were rapidly entrained in the upwardly spiraling gas flow. Changes in injection parameters, including increasing the number of injectors, or increasing the injection velocity did not visibly improve dispersion. A more quantitative evaluation of dispersion was obtained by injecting methane as a tracer gas. Tracer concentrations were then measured at the boiler cross-section in the horizontal superheater passage at a location approximating the midpoint of the sulfation reaction. Figures 6 and 7 compare the dispersion patterns obtained for injection with one and two nozzles. Although the tracer measurements show different patterns, overall dispersion with both configurations is surprisingly uniform.

Results of the tracer measurements were also used to calculate the effect of the dispersion patterns on SO_2 reductions, based upon a relationship of Ca/S ratio to SO_2 capture developed in bench scale tests with near optimum mixing. Results of this analysis are shown in Figure 8, and indicate that varying the injection parameters is not expected to measurably affect SO_2 removal. This result suggests that adequate sorbent dispersion was obtained with each of the injection configuration, and that the effects of mixing become apparent only at the higher Ca/S ratios.

The reactivity of the sorbents from the Tiefstack test was evaluated in a 30 kW thermal (100,000 Btu/hr) test furnace. A wide range of limestone and hydrated lime sorbents have been evaluated in this furnace, providing a data base from which relative reactivity parameters can be determined. Figure 9 shows the test results compared with the range measured for high calcium limestones and atmospheric hydrates. Furnace operating conditions were the same as used in previous studies to allow a direct comparison of sorbent reactivity. The reactivity range of limestone is not as broad as that of hydrates. For both Tiefstack sorbents, however, the reactivity appears to be near the lower end of the range previously measured. During the field test, large particles, in the range of 1000 micrometers, were

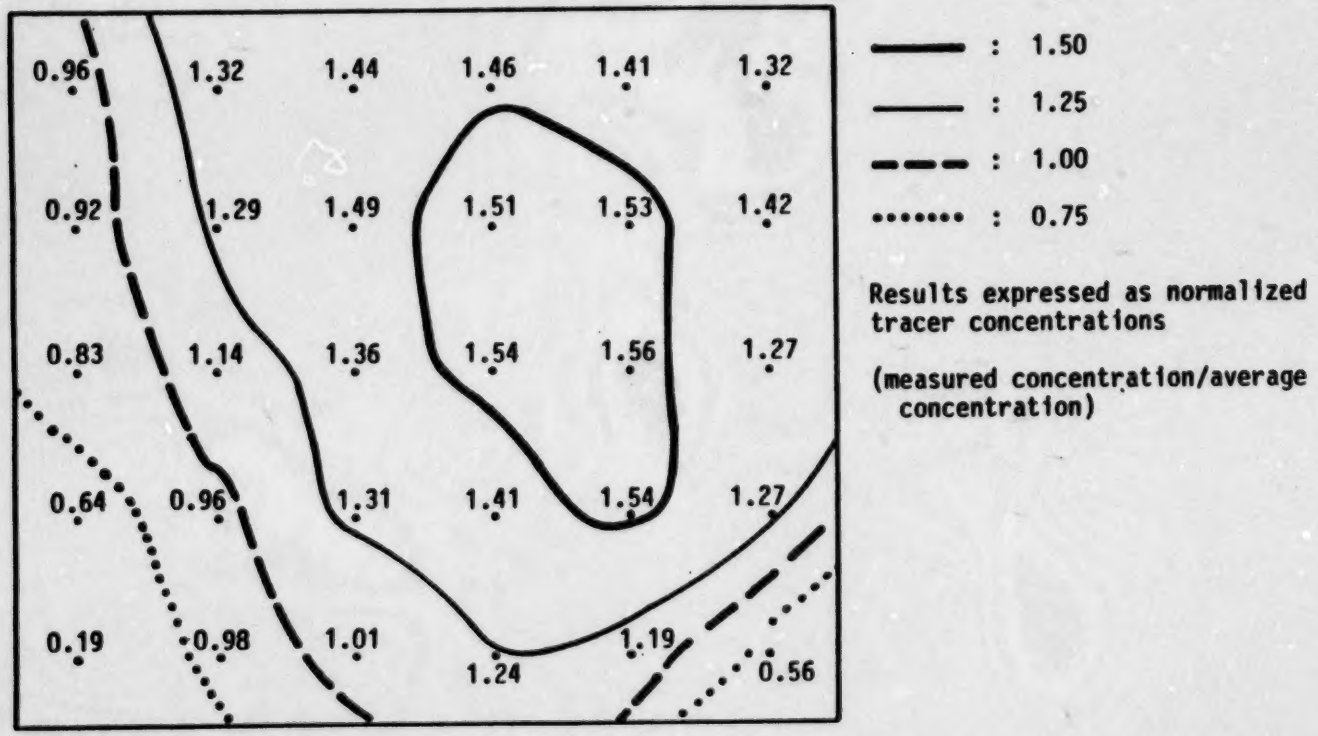


Figure 6. One-nozzle dispersion pattern.

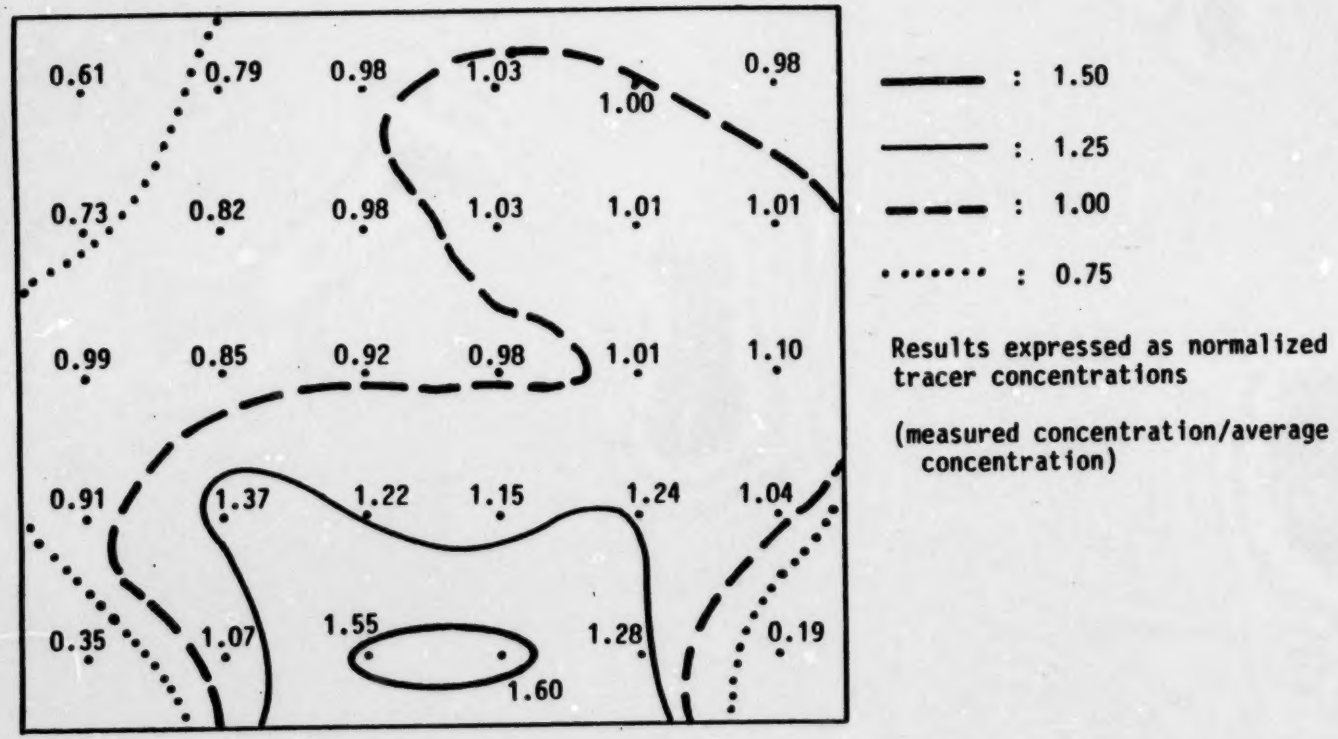


Figure 7. Two-nozzle dispersion pattern.

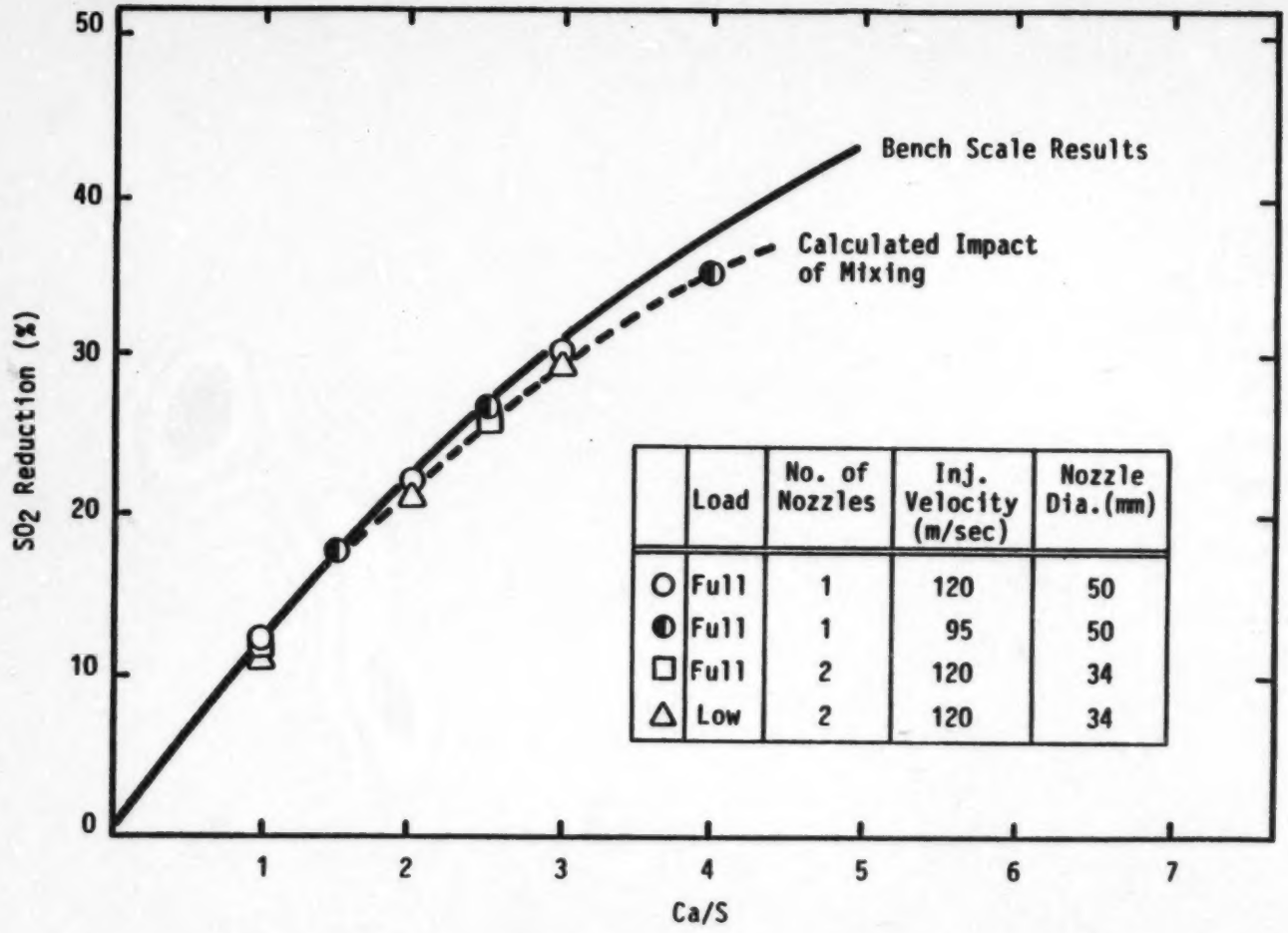


Figure 8. Tiefstack - calculated impact of mixing on SO_2 capture, based on isothermal analysis.

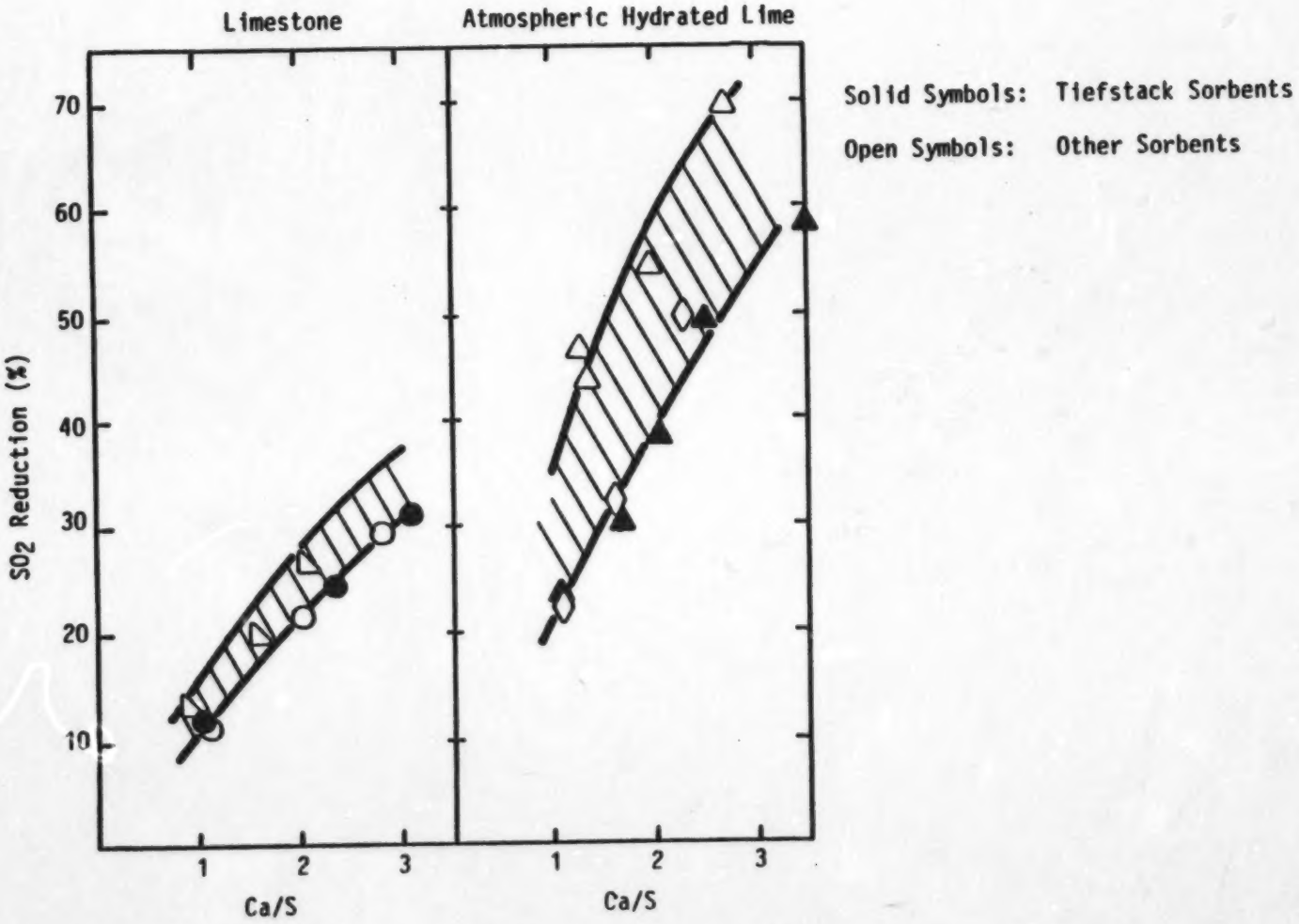


Figure 9. Tiefstack - reactivities of sorbents.

noted in the sorbent. These particles were observed to be propelled all the way across the furnace. The reactivity results and field observations results suggest that improved capture could be obtained at Tiefstack with a reduction in limestone particle size, or the use of commercially available sorbents with higher reactivity.

A mathematical model of the sulfation process was subsequently used in the evaluation of the Tiefstack test results. In its current form, the sulfation model is semi-empirical in nature, in that sorbent reactivity parameters are derived from bench scale data, and the model is then applied to extrapolate to different conditions of injection temperature and quench rate. Since the bench scale test conditions were comparable to those in the full scale boiler, the model quite accurately reflects the actual measurement data and the trends observed. The application was therefore not considered to be an adequate test of the sulfation modeling procedure.

CONCLUSIONS

Measurements of SO_2 capture have been made during sorbent injection tests on Unit 6 at the Tiefstack power plant. The results indicate SO_2 removal rates of 22 percent and 43 percent at a Ca/S ratio of 2, with high calcium limestone and a calcitic atmospheric hydrate, respectively. In view of the apparently low quench rate on this unit, these results were considered to be somewhat disappointing. However, evaluation of the data suggests that the results are, in fact, consistent with bench-scale data, and with our current understanding of process controlling parameters. Temperature measurement on the boiler, supported by heat transfer modeling, confirmed a quench rate of approximately 140K/sec (250 F^{o}/sec), and a temperature at the sorbent injection elevation of 1520K (2280 ^{o}F), which is considered to be close to optimum. Also, contrary to initial expectations, sorbent/flue gas mixing with the one- and two-nozzle injection configurations employed, had little impact on SO_2 removal rates. The boiler data were found to be relatively insensitive to variations in injection parameters. Additionally, small-scale isothermal model tests indicated adequate sorbent dispersion for all configurations studies. Bench-scale sorbent testing under comparable conditions of injection temperature and quench rate yielded SO_2 capture data close to those obtained on the boiler. These data indicate also that the reactivity of the test sorbents is low compared to other similar commercially available materials. The use of alternate sorbents could be expected to increase SO_2 removal at Tiefstack to approximately 30 percent and 60 percent at a Ca/S ratio of 2, for limestone and calcitic atmospheric hydrate, respectively.